Application of a Direct Method in Surface X-ray Crystallography

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Introduction: The electron density of the near-surface region of a crystal could be recovered from x-ray scattering data if the phases of the scattered radiation were known. In the case of surface scattering, the diffracted intensities arise from the square of a linear combination of bulk and surface amplitudes. Since the phases and amplitudes of the bulk component can be calculated accurately, it is possible to use the bulk scattering as a reference wave, and to thereby estimate the phases of the surface component, in analogy with holography. We have developed an algorithm based on the Maximum Entropy Method to estimate the most likely set of surface phases, and to thereby recover the electron density of the near-surface region.

Methods and Materials: Briefly, the algorithm uses Bayesian (or conditional) statistics to select the most likely electron distribution (i.e., surface structure) that can describe a set of surface scattering data. The usual goodness-of-fit criterion (R-factor or chi-squared analysis) must be balanced against a criterion that measures the probability of such an electron distribution occurring. This latter probability is proportional to the number of ways the distribution can occur. (The logarithm of the number of combinations that give rise to an electron distribution is known as the "entropy" of the distribution in analogy to statistical mechanics. Hence, the method is known as the "Maximum Entropy Method".)

Results: On extensive tests using simulated scattering intensities from known surface structures, the method was able to recover the expected surface electron distribution. We note that since the method essentially involves extracting electron density by an inverse Fourier transform of a set of structure factors (without knowledge of their phases), it is imperative that a large set of structure factors, extending to as large an out-of-plane momentum transfer as possible, be collected for analysis. Experiments to collect such a data set were carried out at beamline X16A, whose geometry allows estimable out-of-plane momentum transfers to be reached.

We have made the initial attempts to apply this method on (2x1)-reconstructed Ge(001) surfaces, whose essential structural features are known. The method has been able to reproduce electron density around the expected bulk-like atomic positions. At this preliminary stage, a reliable extraction of density at the expected reconstructed positions is still forthcoming.

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